LONG –TERM VARIATIONS (2000-2010) OF GROSS ALPHA, GROSS BETA AND GAMMA RADIONUCLIDES IN SURFACE AIR: ANALYSIS OF THEIR VARIATION PREDICTION MODEL AND DOSES

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In our laboratory, we are continuously measuring the radioactivity in the surroundings as part of a general project undertaken by many laboratories in Spain in collaboration with the Spanish Nuclear Security Council (CSN). In this paper gross alpha, gross beta and gamma emitting radionuclides are routinely measured in samples of airborne dust samples and our sampling site is geographically far from the influence of nuclear installations. The sampling site is one of the environmental radioactivity monitoring network stations operated by the CSN, under cooperative agreement with the University of Málaga through the Environmental Radioactivity Research Group. The sampling point was located 12 m above the ground, on the roof of the Faculty of Sciences, University of Málaga. The site where the measurements were carried out (4º 28′ 8″ W; 36º 43′ 40″ N) is in the North-West of the city, 5 km away from the coastline. Málaga is the mayor coastal city of Andalusia region, South Spain. The Spanish city on the Mediterranean is distinguished by its mild, temperate and warm climate with low rainfall (550 $mm \cdot yr^{-1}$) and around 320 days of sun a year. All detected gamma radionuclides are of natural origin. We analyze the time series of gross alpha, gross beta and gamma radionuclides composition of air. The data are sufficiently numerous to allow us to examine variation in time and through these measurements we have established several parameters that should be of importance in understanding any trends in radionuclide concentrations in the atmosphere. Seasonal variations of gross alpha, gross beta activities and radionuclides concentrations in the atmosphere show a tendency for a maximum in the spring and summer and a minimum in autumn and winter. In this work, the data on concentrations and meteorological data have been made use of in order to determine a model for gross alpha, gross beta and gamma radionuclides. An ARIMA model was used to predict the gross alpha and gross beta activities .A satisfactory agreement between the results of the model and the measurements was highlighted. Also we have estimated the doses due to the radionuclides in the air aerosols from air inhalation in Málaga.

INTRODUCTION

Radioactivity in the atmosphere originates from natural radioactive decay, cosmogenic production and from nuclear weapons testing and nuclear accidents. Inhalation is one of the pathways by which radioactive nuclides are incorporated into the human body .Monitoring of atmospheric radionuclides is an important part of avoiding or eliminating the risk of disease to the general public should there be an emergency. Several sets of measurements have been performed in order to a achieve a better understanding of this subject (Papastefanou, 1994; Gómez et al., 1996, Dueñas et al.,2004).

In this paper, gross alpha, gross beta, ^{7}Be and ^{210}Pb activities were routinely measured in samples of airborne dust samples during 11 year period (2000-2010). Using these data, the present research was undertaken with the following principal goals:

1. To perceive the variations of gross alpha, gross beta, ^{7}Be and ^{210}Pb activities deriving the statistical estimates characterizing their distributions and analyzing the harmonic components of the data set.
2. To model the data by ARIMA models. These models are fitted to time series data either to better understand the data or to predict future points in the series (forecasting). The model is generally referred to as an ARIMA(p,d,q) model where p, d, and q are non-negative integers that refer to the order of the autoregressive, integrated, and moving average parts of the model respectively.

3. To validate the previous model we performed a study applying the equation obtained to data which were not used in the analysis of the models.

4. To estimate the doses to the ^{7}Be and ^{210}Pb radionuclides in the air aerosols from air inhalation.

MATERIAL AND METHODS

Airborne dust samples were collected at a height of 12 m above the ground in Málaga (36º 43″ 40″ N; 4º 28′ 8″ W). Aerosol samples were collected weekly in cellulose membrane filters of 0.8 µm pore size with an air flow rate of approximately 40 l min-^{1}. The air sample was lodged in an all weather sampling station. The gross-α activities were measured by an α-scintillation detector ZnS(Ag).The alpha particle counting system consisted of a 50 mm diameter phototube and the classic elements of a counting chain. The alpha particle detector was a mylar sheet having the same diameter as the phototube and a homogeneous layer of ZnS(Ag) crystals. The gross β activities was measured by a low level alpha–beta counting system which consisted of a low background multiple detector having four sample detectors (CANBERRA HT-1000). The sample detectors were gas flow window-type counters, of 5 cm diameter. Detailed description and calibration of the proportional counter is given in Dueñas et al., (1997) Measurements by gamma–spectrometry were performed to determine the ^{210}Pb and ^{7}Be activities of the samples using an intrinsic REGe detector. A monthly composite sample containing 4-5 filters was formed (average volume 1600 m^{3}) for the ^{210}Pb and ^{7}Be determination. The errors reported are propagated errors arising from the one sigma counting uncertainty due to detector calibration and background correction. The concentrations were corrected for decay to the mid-collection period. Details of the low-background gamma-ray detection system used and the calibration for gamma spectrometry have been previously described (Dueñas et al., 1999).

RESULTS AND DISCUSSION

Statistical analyse and uncertainties of measurements.

Table 1 summarises the main statistical variables calculated for the gross alpha and gross beta activities as well as for the ^{7}Be and ^{210}Pb concentrations over the whole sampling period: 1 January 2000 until 31 December 2010. All activities are given in Bq m^{-3}. The measurements were performed once the short-lived radon daughter had decayed. The artificial radionuclides investigated were below the minimum detectable activity (MDA).

	Gross α (Bq m^{-3})	Gross β (Bq m^{-3})	^{7}Be (Bq m^{-3})	^{210}Pb (Bq m^{-3})
N of cases	132	132	132	132
Arit. mean	5.80E-05	6.70E-04	4.40E-03	4.80E-04
Geo. mean	5.60E-05	6.20E-04	4.00E-03	4.10E-04
Maximum	11.00E-05	15.00E-04	15.00E-03	14.00E-04
Minimum	1.70E-05	0.01E-04	0.50E-03	0.40E-04
Std. deviation	1.50E-05	2.20E-04	1.70E-03	2.40E-04
Std error	0.10E-05	0.20E-04	0.16E-03	0.20E-04
Variation coefficient	26 %	32%	39%	51%
Skewness (GI)	2.1	2..2	6.3	2.7

Table 1. Statistical summary of the radiometric data collected from January 2000 until December 2010.

Due to the value of skewness (GI), the gross alpha, the gross beta activities and specific activities of ^{7}Be and ^{210}Pb in air should fit approximately to log normal distribution. The geometric mean was used in further analysis and comparisons as the main characterization factor for the gross alpha, gross beta and specific activities of ^{7}Be and ^{210}Pb.

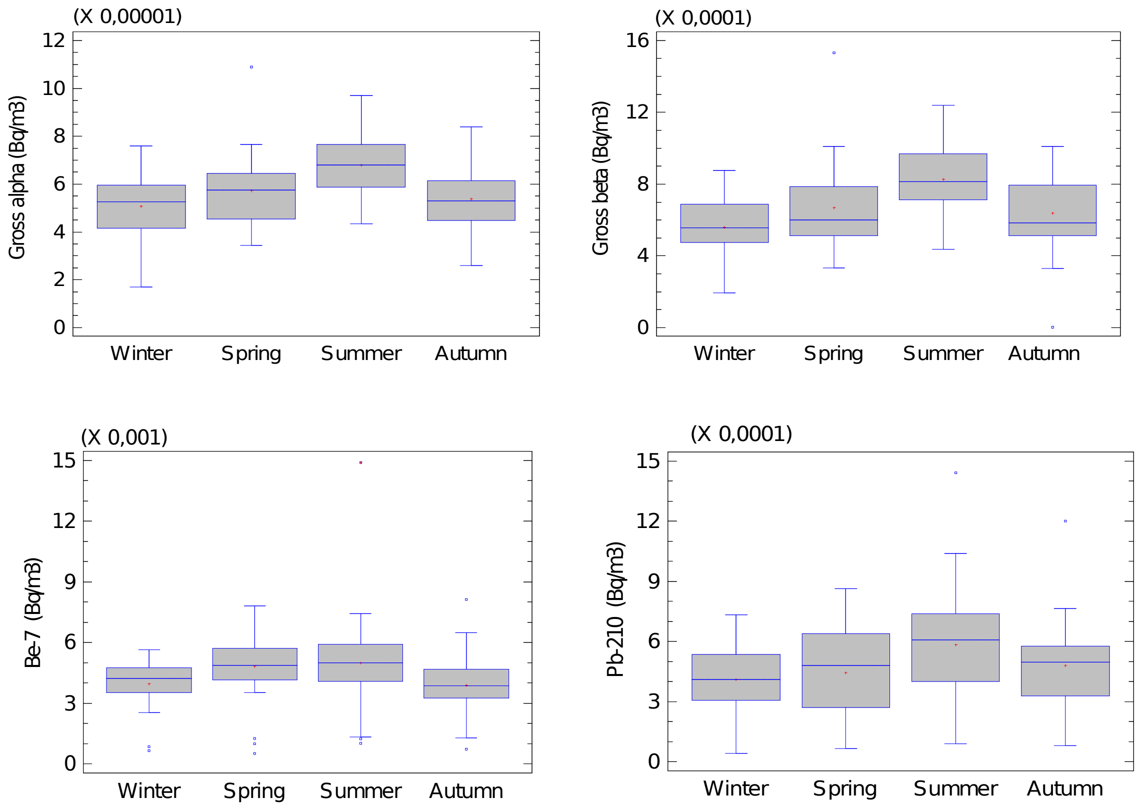

Figure 1. Seasonal variation of gross alpha, gross beta, ^{7}Be and ^{210}Pb activities.

Figure1 show the box and whisker diagrams for monthly of gross alpha, gross beta and of ^{7}Be and ^{210}Pb in air activities. All activities show a marked monthly variation with higher values of activity in summer and spring months and lower values in other months. The results of fig. 1 indicate significant differences in the

activity levels of ^{7}Be not only between summer and the rest of the seasons but also between spring and other seasons. The high activities in the summer months are caused by increased vertical transport of ^{7}Be activities from the upper troposphere due to decreased stability of the troposphere during the summer months. The ^{7}Be concentrations in spring are higher than the activities in autumn and winter and are normally assigned to the thinning of the tropopause which takes place at midlatitudes, resulting in air exchange between stratosphere-troposphere during late in winter and early spring. In other words, the monthly behavior of the mentioned tracers shows similar evolution during the summer months but a different evolution in other months.

From a visual inspection of the data, seasonal changes seem to be produced and are commonly attributable to different factors such as temperature, atmospheric stability or frequency and amount of precipitation. In order to find the meteorological factors that influence the specific activities of gross alpha, gross beta, ^{7}Be and ^{210}Pb activities, we performed a simple regression with meteorological factors. In our analyses we used monthly average of the temperature expressed in ºC, monthly average of precipitation expressed in mm, monthly average of the relative humidity expressed in % and monthly average wind speed (expressed in m/s). Table 2 shows the results.

	Gross α	Gross β	^{7}Be	^{210}Pb
Temperature	0.52	0.55	0.34	0.30
Precipitation	-0.38	-0.37	-0.19	-0.21
Relative humidity	-0.31	-0.29	-0.20	-0.21
Wind speed	-0.12	-0.10	-0.12	-0.09

Table 2. Linear correlation coefficients between gross alpha, gross beta, ^{7}Be , ^{210}Pb activities and some meteorological factors.

Judging from the linear coefficient values, it can be said that the gross alpha, gross beta, ^{7}Be and ^{210}Pb activities present pronounced positive correlation with the average monthly temperature of air, a weak negative correlation with the average speed of wind and a negative one with the other meteorological factors (relative humidity and amount of precipitation).The coefficients of correlation range between -0.09 and 0.55 indicating relatively weak relationship between the variables. The weak relationship in this study suggests that gross alpha, gross beta ^{7}Be and ^{210}Pb activities at our site might be controlled by the interactions of complex processes, such as source and scavenging intensity .Based on the available data and the low dependencies found between the different variables, no predictive model could be built.

ARIMA model

A stochastic model based on studying the time series has been proposed First, to study the temporal behaviour and the harmonic components of the data sets were analyzed .We obtained a periodogram based on a Fourier analysis which assumes that the data time series were formed by the superposition of sinusoidal components of different frequencies for gross alpha, gross beta, ^{7}Be and ^{210} Pb activities. The intensity of the periodogram, I(υi), was defined as:

$$I(\upsilon_i) = \frac{2}{N}\left\{\left[\sum_{t=1}^{t=N} A_\alpha(t)\cos 2\pi\upsilon_i t\right]^2 + \left[\sum_{t=1}^{t=N} A_\alpha(t)\sin 2\pi\upsilon_i t\right]^2\right\}$$

with i=1,2,3…q, where $q=(N-1)/2$ for odd N and $q=N/2$ for even N. The periodogram was then the plot of $\boldsymbol{I}(\upsilon_i)$ against υ_i, where $\upsilon_i=i/N$ is the ith harmonic of the fundamental frequency $1/N$, up to the Nyquist frequency of 0.5 cycles per

sampling interval. Since $I(\upsilon_i)$ was obtained by multiplying $A_\alpha(t)$ and $A_\beta(t)$ by sine and cosine functions of the harmonic frequency, it will take on relatively large values when this frequency occurs in $A_\alpha(t)$ and $A_\beta(t)$. As a result, the periodogram maps of the spectral content of the series, indicate how its relative power varies over the range of frequencies between 0 and 0.6 (Hewit, 1992). Figure 2 shows the periodograms corresponding to the gross alpha, gross beta, ^{7}Be and ^{210}Pb activities.

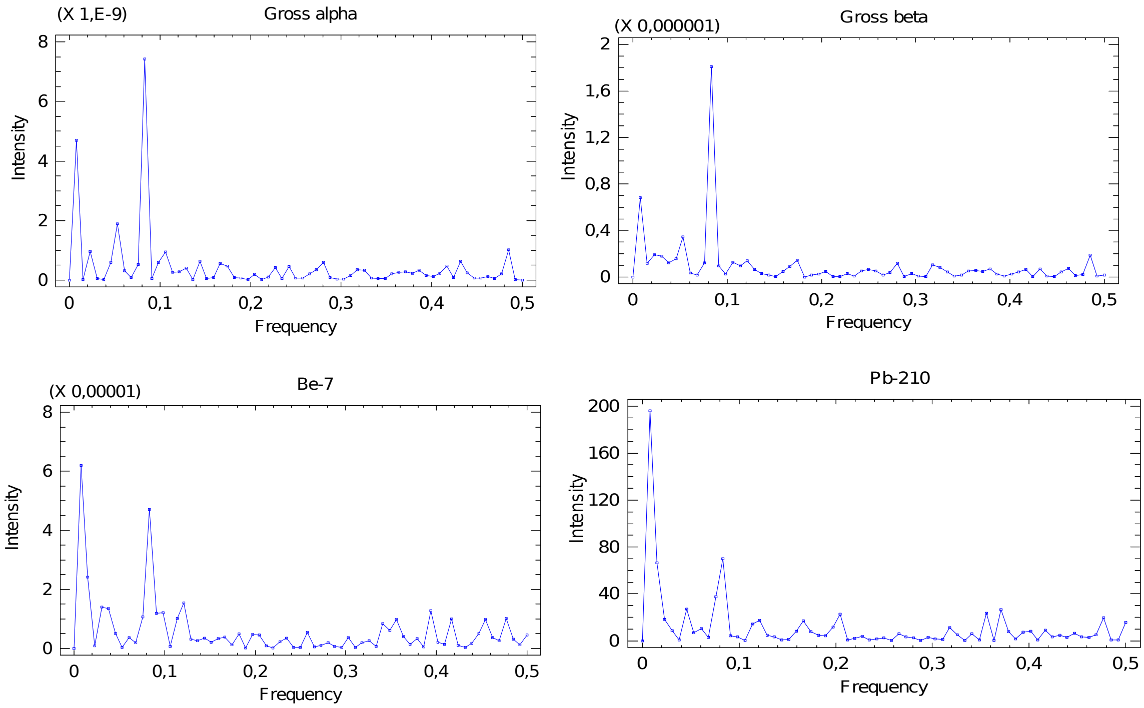

Figure 2. Periodogram corresponding to the gross alpha, gross beta, ^{7}Be and ^{210} Pb activities.

One observes large peaks for periodograms of gross alpha and beta activities, with frequencies of 0.08 month^{-1} which correspond to periods of 12 months .This indicate that there are cyclical phenomena with a period of approximately 1 year. The periodograms of ^{7}Be and ^{210}Pb activities are plus complexes and for this reason we are not developed the ARIMA model for both radioisotopes. In order to know if the time series of gross alpha and gross beta are suitable to stochastic model, we would study two functions, the autocorrelation function (ACF) and the partial autocorrelation function (PACP).

These functions has been evaluated and the ARIMA model estimated are ARIMA(1,0,0)x(2,1,2)12 with constant to gross alpha activities and ARIMA(2,0,0)x$(0,1,1)_{12}$ to gross beta activities.

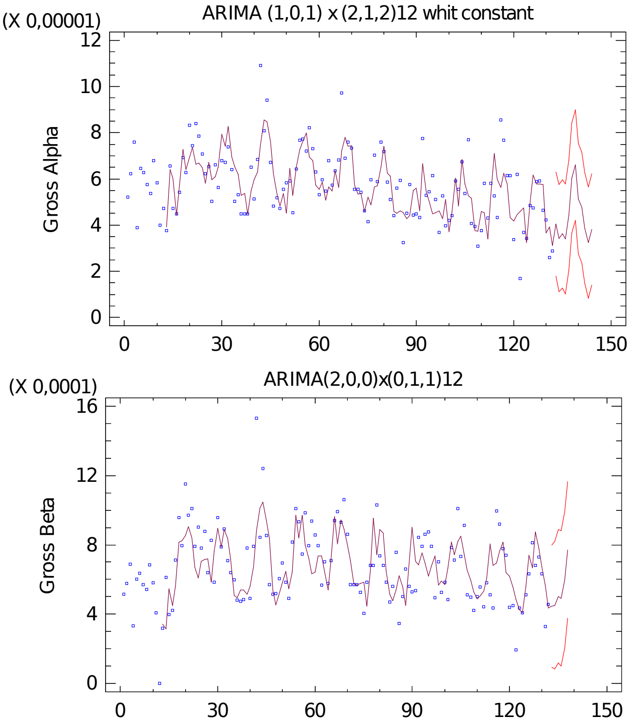

Figure 3. ARIMA(1,0,1)x(2,1,2)$_{12}$ with costant to gross alpha activities and ARIMA(2,0,0)x(0,1,1)$_{12}$ to gross beta activities.

To test the validity of these models in the estimation of gross alpha and gross beta activities, we performed a study by applied the model ARIMA(1,0,1)x(2,1,2)$_{12}$ with costant to gross alpha activities and ARIMA(2,0,0)x(0,1,1)$_{12}$ to gross beta activities to monthly data which have not yet been used to obtain the equations of the ARIMA models. To data correspond to the last 12 months of the year 2010.The study was conducted by comparing the activities predicted by ARIMA models to those experimentally obtained. In figure 4, the predicted data versus experimentally data with the corresponding prediction limits at 95% confidence level were plotted

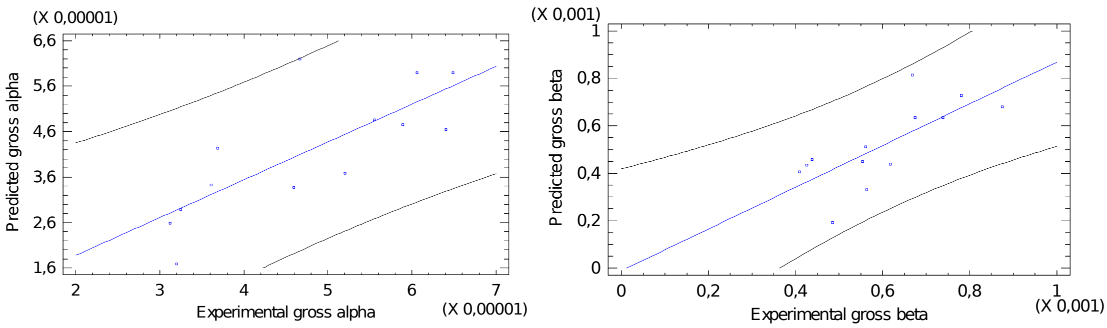

Figure 4. Predicted by the ARIMA(1,0,1)x(2,1,2)$_{12}$ with costant to gross alpha activities and ARIMA(2,0,0)x(0,1,1)$_{12}$ versus experimental values with the prediction limits at 95% confidence level.

The R-squared statistic indicates that the model as fitted explains 60% and 54% of variability in the experimental data for gross alpha and gross beta respectively.

Dosimetry

We have estimated the doses due to the radionuclides ^{7}Be and ^{210}Pb in the air aerosols from air inhalation in Málaga. The used formulate has been: Effective annual dose for adult = (arithmetic mean of radionuclide (Bq/m^3) x Inhale air (m^3/year) x Conversion factor (Sv/Bq)/1000mSv^{-1} .We have supposed that an adult inhale 84096 m^3 /year. The conversion factor has been 4.3x10^{-11} Sv/Bq for ^{7}Be and 1.1x10^{-6} ^{210}Pb Sv/Bq for The Annual effective dose rate received by the adult was1.6x10^{-5} mSv/y for ^{7}Be and 4.4x10^{-2} mSv/y for ^{210}Pb (RD 783/2001). Also, we can conclude that the radiation dose to adult resulting from the ^{7}Be and ^{210}Pb in the air aerosols is negligible compared to the average annual dose from natural sources(2,4 mSv/y).

Conclusions

1. The results obtained for airborne particle samples measured during 11 years for the gross alpha, gross beta, ^{7}Be and ^{210}Pb activities give geometric means of 5,6x10^{-5}, 6,2x10^{-4}, 4x10^{-3} and 4.1x10^{-4} Bq/m^3 respectively.

2. The correlation coefficients between gross alpha, gross beta ^{7}Be and ^{210}Pb and some meteorological parameters range between -0.09 and 0.55 indicating a relatively weak relationship between the variables, this is the reason because no predictive model could be built.

3. The periodograms corresponding to gross alpha and gross beta activities shows that there are cyclical phenomena with periods of approximately 1 year.

4. We development two Auto-Regressive Integrated Moving Average (ARIMA) models, ARIMA $(1,0,1)x(2,1,2)_{12}$ with costant to gross alpha activities and ARIMA$(2,0,0)x(0,1,1)_{12}$ to gross beta activities.

5. The R-squared statistic to test the validity of the ARIMA models is greater for gross alpha than gross beta activities.

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